

Analyse der Defektausheilung in einkristallinen Goldschichten nach Bestrahlung mit Goldionen

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Analysis of Defect Annealing in Monocrystalline Gold Foils after Gold Ion Irradiation

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Monocrystalline gold foils were irradiated at -196°C with gold ions parallel to $[101]$ or at random incidence. Defect production and subsequent isochronal recovery between -180 and 40°C were examined by means of the electrical resistance for energies from 5 to 30 keV and doses from 5×10^{12} to $5 \times 10^{15}\text{ ions/cm}^2$. The annealing spectra are characterized by an intense dose-dependent peak between -40 and -25°C which is interpreted as being due to the recombination of freely migrating interstitials ($E_{11}^M = (0.77 \pm 0.07)\text{ eV}$) with vacancies. After high-dose preirradiation and subsequent annealing the recovery in the dominant peak is reduced in favour of a new peak appearing between 0 and 40°C . This is explained in terms of a migration of vacancies to defect clusters.

Einkristalline Goldschichten wurden bei -196°C mit Goldionen parallel zu $[101]$ bzw. ungerichtet bestrahlt. Die Defektproduktion und die anschließende isochrone Erholung zwischen -180 und 40°C wurden für Energien zwischen 5 und 30 keV und Dosen zwischen 5×10^{12} und $5 \times 10^{15}\text{ Ionen/cm}^2$ mit Hilfe des elektrischen Widerstands untersucht. Das Erholungsverhalten wird geprägt durch ein intensives dosisabhängiges Maximum zwischen -40 und -25°C . Dieses kann der Rekombination frei wandernder Zwischengitteratome ($E_{11}^M = (0.77 \pm 0.07)\text{ eV}$) mit Leerstellen zugeschrieben werden. Nach starker Vorbestrahlung und anschließender Ausheilung sinkt die Erholung in dem ausgeprägten Maximum zugunsten eines neu auftretenden Maximums zwischen 0 und 40°C . Dieses wird gedeutet als Wanderung von Leerstellen zu Defektagglomeraten.

1. Introduction

In spite of nearly twenty years of intense research, a generally accepted assignment of the recovery stages [1], observed after low-temperature irradiation of f.c.c. metals, to the migration and recombination of various point defects has not yet been obtained [2]. Two models have been proposed [3, 4] which differ mainly by assuming the free migration of the most simple interstitial configuration either below about 50°K (stage I_E, migration energy $E_{11}^M \approx 0.1\text{ eV}$) [3] or above about 200°K (stage III, $E_{11}^M > 0.5\text{ eV}$) [4]. Both models contain free parameters enough to explain all experimental results under more or less reasonable assumptions.

Interstitials possess a much higher formation energy than vacancies so that their properties, unlike those of vacancies [2, 5], cannot be deduced from equilibrium measurements at high temperatures or non-equilibrium experiments after quenching. Excess concentrations of interstitials may be produced only by

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irradiation with ions, usually of the same kind as the target material. In principle, there are two possibilities:

(i) Upon irradiation with low-energy ions (some 100 eV) target interstitials may be carried into the specimen via replacement collision sequences along closely packed lattice rows ('dynamic crowdions'). These interstitials are thought to come to rest at least some 10 or perhaps nearly 1000 atomic distances below the surface [6].

(ii) Upon injection parallel to low-index directions ions of an energy higher than some keV may execute correlated collision sequences with large impact parameters along the atomic rows ('channelling'). Well-channelled projectiles are thought to be stopped mainly via inelastic electronic collisions and to come to rest as interstitials without producing Frenkel defects. Dechannelled ions on the other hand cause defect cascades.

The object of this paper is the question whether channelling allows the production of an excess concentration of interstitials. In addition, the migration parameters of interstitials are investigated.

2. Experimental

The projected experiments required an apparatus which allowed the gold foils to be annealed in situ after irradiation at -196°C . The experimental set-up is shown schematically in Fig. 1. The construction elements (ion source (1) with extraction electrode (2) and annealing device with acceleration electrode (3)) were mounted on the parallel ends of a sphereflange-T-glassvessel (4) (DURAN 50, Schott & Gen.). During operation of the ion source the vacuum was better than 2×10^{-6} Torr.

2.1 Ion source

The metal ion source was constructed according to the design of Magnuson et al. [7]. The operating conditions were determined with gold as a source material (ionization voltage $U_i = 9.2$ V). The gold vapour pressure was kept at the optimum value of about 10^{-3} Torr (only 300 W were needed to maintain the oven temperature at 1500°K). A magnetic field of at least 20 Oe were necessary to produce a stable plasma.

In order to obtain a sufficiently pure ion beam the maximum ionization voltage used for the irradiation experiments was 12 V. Thus it was ensured that the ion beam was kept free from rest gases, especially oxygen ($U_i = 12.1$ V) and nitrogen (15.7 V). In spite of these precautions there was a 'zero-current' of some nA during the operation of the ion source without gold. By means of the high secondary electron yield measured at the target the charged particles could be identified as light ions. These probably originated from

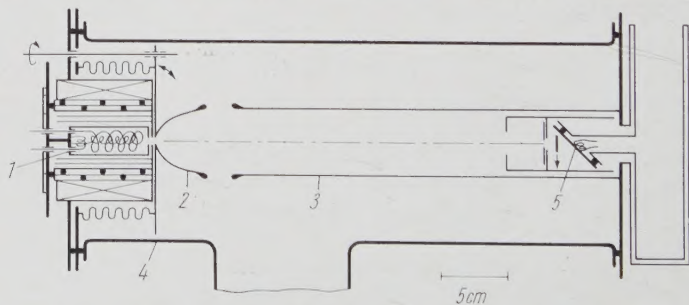


Fig. 1. Experimental equipment for irradiation and annealing of the gold foils. 1 — ion source, 2 — extraction electrode, 3 — acceleration electrode, 4 — glass vessel, 5 — heating plate (sample position)

(Na,K) impurities of the alumina tubes (DEGUSSIT Al 23, Degussa) which served as insulating material for the electrical lead-ins of the heating filament and the cathode. The irradiation was normally performed with an ion current of some μA (beam diameter about 3 mm) so that the fraction of impurities was not larger than 10^{-3} .

2.2 Acceleration equipment

The ions produced by electron collision were drawn off the plasma surface (potential U_p) at the 3 mm oven exit by means of the extraction electrode (U_1). The ions achieved their final energy in the acceleration tube (U_2). The exact adjustment of the beam to the respective target site could be attained by adequately tilting the extraction electrode. The beam quality was determined mainly by the acceleration stage. The geometry of that immersion lens was found from the ion-optical data of two long cylindrical electrodes (U_0 , U_2). Extremely low values of U_0/U_2 were needed to avoid noticeable aperture defects [8]. The potential fields and the focal points for this case were calculated by Biersack [9]. A suitable lens geometry could be compiled from the existing data.

The 3 mm entrance window of the extraction electrode coincided with the low-energy focus F_1 and its metallic surface was shaped to follow the potential plane U_1 through F_1 . Extraction potential and plasma density could easily be adjusted to shift the 'cross-over' of the extraction stage to the site of F_1 . By measuring the beam profile with a specially designed probe the divergence of the ion beam was found to be less than 0.1° for $U_1/U_2 = 1/60$. This result agreed with the calculated values within the experimental uncertainty of a few per cent. A space-charge-like increase of the maximum beam current ($i_m \sim U_1^{3/2}$) was found for a fixed position of the 'cross-over', e.g. $i_m = 120 \mu\text{A Au}^+$ for $U_1 = 1 \text{ keV}$.

2.3 Annealing device

The main part of the annealing device (Fig. 1) was a heating plate (5) tilted by 45° to the beam direction. The aluminium plate oxidized on the vacuum side served as a heat transmitter for the specimens. It was mounted at the end of a copper tube connected to a container for liquid nitrogen. Vacuum sealing as well as thermal contact between aluminium plate and copper tube was achieved with indium-O-rings. The ion beam was directed towards the irradiated sample through a suitable aperture. A mechanical shutter served to control the ion current and the irradiation dose. During irradiation and measurement of resistance changes the aluminium plate was in direct contact with liquid nitrogen. Thermal annealing was done in situ by means of a heating coil (1 Nc I 10, Philips) fixed to the backside of the aluminium plate. The maximum heating and cooling rates were 2°C/s .

In order to achieve the necessary electrical insulation between the gold foils and the heating plate and simultaneously to provide sufficient thermal contact the glossy polished aluminium surface was oxidized. This was done after Walkenhorst [10] by anodic oxidation with an electrolyte of a 3% solution of diammoniumhydrogencitrate $[(\text{NH}_4)_2\text{C}_6\text{H}_6\text{O}_7]$ in water. It turned out that a porefree alumina layer could only be obtained by slowly increasing the oxidation voltage. For the maximum voltage of 300 V, applied in steps of 20 V (total oxidation time about 3 h), the thickness of the alumina layer should be 2650 Å according to Davies et al. [11].

2.4 Preparation of gold foils

Monocrystalline gold foils of (100) orientation, $0.3 \times 6 \text{ mm}^2$ and 2000 to 3000 Å thick, were prepared by evaporation of 99.999% pure gold (Degussa)

through masks onto (100) cleavage planes of KI or NaCl [12] in a vacuum better than 10^{-6} Torr. The foil thickness was determined by Tolansky interferometry [13]. In order to control the quality of the foils 200 keV electron microscope diffraction and transmission studies were carried out. The density of crystal defects, which are mainly stacking faults and twins, was about $2 \times 10^9/\text{cm}^2$.

Two foils for each experiment (one sample for irradiation and another for reference) were removed from the substrate in distilled water and then attached to the heating plate. The specimens had to adhere completely flat to the base plate after the first drying process because later attempts for correction via repeated detaching and drying did not result in a smoother film. The extreme fragility of the foils did not allow direct fixing of electrical lead-ins to the specimens. Therefore contact stripes, some microns in thickness, were evaporated onto the ends of the samples. They were connected to the current and the potential leads by means of a low melting solder (40% Bi, 25% Pb, 15% Hg, 10% Sn, and 10% Cd [14]). The sample temperature was detected by means of a coated miniature thermocouple (2 AB Ac 05, Philips) which was fixed to the heating plate near the foils. The resistance of the irradiated and annealed sample was measured at -196°C with respect to the reference probe thereby avoiding the effect of small temperature fluctuations on the base plate. Applying a potentiometric technique the resistivity could be detected to within $3 \times 10^{-11} \Omega \text{ cm}$.

2.5 Electronic temperature control

In order to get an exact information about the recovery as a function of temperature well-defined temperature-time runs were performed. The temperature regulation was obtained with a proportional amplifier using the reference probe as a control junction. As expected the resistance of the reference probe depended linearly on temperature so that a given temperature could easily be achieved after calibration. Due to the limited rise of the control curve, the temperature coefficient of input-offset voltage of the operational amplifier, and the possible errors in determination of thermopower the maximum uncertainty in temperature during isochronal annealing was $\pm 0.3^\circ\text{C}$.

3. Experimental Results and Interpretation

3.1 General remarks

When analysing defect production and recovery curves one has to keep in mind that electrical resistance measurements cannot distinguish between different types of defects. In this case additional influences of foil thickness (size effect), inhomogeneity of defect distribution, and sputtering have to be taken into account.

In foils of a thickness t of the order of the mean free path λ of the electrons in the bulk material the resistivity ϱ_F may considerably exceed the bulk value ϱ if the electrons suffer inelastic (diffuse) reflections at the surfaces. For $\varrho \sim 1/\lambda$ one has approximately $\varrho_F = \varrho + (1 - p) k/t$ [15], k being a constant number and p the fraction of electrons elastically scattered at the surfaces. Assuming that p approaches a constant value even for small irradiation doses one gets $\partial\varrho_F/\partial\varrho = 1$.

Even if the size effect is ignored one cannot expect a resistance change proportional to the integral defect concentration c because the measurements performed parallel to the irradiated surface yield only an average resistivity $\bar{\varrho}$.

It may be shown, however, that for the resistance changes observed $\partial\bar{\rho}/\partial c$ is approximately constant.

Sputtering has to be considered when analysing the defect production and the recovery spectra in the high-dose case. From measurements of other authors [16, 17] one may estimate that for a 30 keV gold ion irradiation parallel to [101] and 45° oblique to the surface the sputtering yield γ roughly equals 25. Our own experiments on resistance changes of 3000 Å foils after repeated intense bombardment yielded a value of $\gamma = 20 \pm 5$.

3.2 Defect production

The defect production was examined at an ion energy of 30 keV through three orders of magnitude of irradiation dosage from 5×10^{12} to 5×10^{15} Au⁺/cm². For lower doses the resistance changes were so small that the shape of the recovery spectrum could not be detected with sufficient accuracy. It was found (Fig. 2) that the magnitude of the resistance changes other than the dose dependence is strongly determined by foil orientation and pretreatment. The largest resistance is found for irradiation of virgin gold foils parallel to [101]. In the case of random incidence the defect production is smaller by a factor of three. An effect of that amount has already been observed by Merkle [18]. In foils strongly preirradiated with a dose Φ_p and annealed up to 40 °C the resistance increase upon bombardment parallel to [101] is smaller than in virgin specimens.

In any case the resistance increases proportionally to the square root of the dose Φ . A $\sqrt{\Phi}$ -dependence of resistivity has also been found for intense 3 MeV electron irradiation of copper foils at 120 °K [19]. Preliminary experiments on gold showed similar results. The observation of vacancy and interstitial clusters in those electron-irradiated foils [20] is in agreement with model calculations of Lück and Sizmann [21]. They could show that spontaneous recombination results in the formation of vacancy and interstitial clusters. By analogy it is tempting to explain the $\sqrt{\Phi}$ -dependence found upon ion bombardment partly by spontaneous recombination. This would mean that even in the case of parallel injection of gold ions vacancies are produced in considerable concentrations.

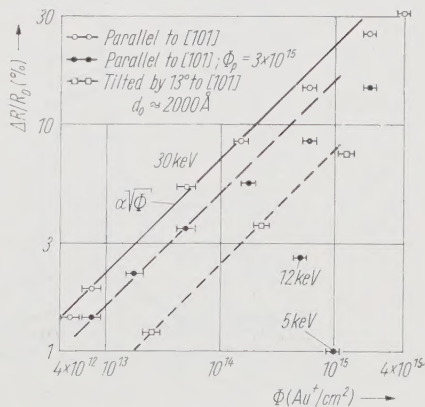
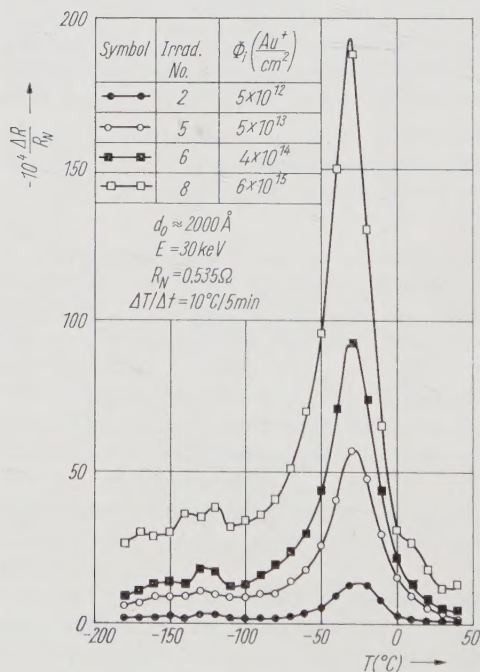


Fig. 2. Fractional resistance increase at -196 °C versus irradiation dose for different foil orientation and pretreatment

For an explanation of the above mentioned results the experiments of Thomas et al. [22] are useful. The authors investigated the defect production in monocrystalline gold foils by aligned bombardment with 3 to 120 keV gold ions at room temperature. As an appropriate measure they used the well-known 'black spots' observed in damaged foils with an electron microscope. These spots could be identified as vacancy clusters and centres of defect cascades. The most important result was that observable defects are even produced far beyond the 'amorphous' range of the injected ions. This can only be explained by dechannelling of originally channelled projectiles. Therefore the hope for a production of an excess concentration of interstitials via channelling has to be abandoned for the present.

With the help of the investigations of Thomas et al. [22] all the details of Fig. 2 may be understood qualitatively. In the case of aligned bombardment the resistance change is larger than for random injection because the greater range of channelled projectiles leads to more cascades at larger depths. The lowering of the defect production in the high-dose-pretreated foils may be explained by the presence of point defect clusters. Vacancy clusters in gold are known to dissolve considerably only above 500 to 700 °C [23]. Therefore, they are stable up to 40 °C and may produce an enhanced amount of spontaneous recombination upon repeated irradiation. In the case of low-energy bombardment the defect production decreases for two reasons: (i) because the range of the projectiles decreases rapidly with energy and (ii) because the number of defect cascades produced per injected ion is also markedly reduced below 15 keV [22].



3.3 Annealing behaviour

After bombardment at -196°C (irradiation parallel to $[101]$ if not indicated otherwise) the monocrystalline foils were annealed isochronally in steps of 10°C per 5 min. Fig. 3 shows some examples of the recovery spectra obtained. The investigations were carried out successively on the same foil and the dose was increased by about one order of magnitude in each case. The resistance changes $-\Delta R/R_N$ (sample: R , reference probe: R_N) are the uncorrected differences of the data measured between two successive annealing steps. The value at -180°C ($\Delta T = 16^\circ \text{C}$) was converted to an annealing step of 10°C .

Fig. 3. Isochronal recovery spectra for enhanced irradiation doses

The characteristic feature of the recovery spectra is the pronounced peak centered at about -30°C . The recovery proceeds almost continuously below -100°C . For low doses a small peak occurs at about -125°C which seems to split for higher doses. The recovery is completed near 40°C , especially for low and medium doses. In what follows the recovery above -80°C is called stage III as in the common nomenclature [1].

The activation energy E characterizing the recovery stage may be determined for one sample at a time by means of a cyclic isochronal annealing program after Bell and Sizmann [24]. The slopes in the inflection points were determined graphically because the extrapolation procedure given in [24] was not successful for experimentally obtained curve shapes. The analysis of all cyclic recovery spectra yielded an activation energy of (0.77 ± 0.07) eV for defect migration in stage III. The error indicated results from uncertainties in temperature control and slope determination. The activation energy is found to be constant in the temperature range -50°C to -10°C . This suggests the existence of a single-activated process.

A more exact analysis of Fig. 3 shows that the maximum in the recovery rate is shifted towards lower temperatures with increasing dosage. This shift of peak position is typical for recovery processes which result from free migration of point defects to their complementary partners and eventual recombination. From the temperature shift ΔT_m of peak position another characteristic of the recovery process, the reaction order n , may be deduced. In Fig. 4 the temperature shifts are shown as a function of the total relative resistance recovery (i irradiation number in Fig. 3). For comparison the temperature shifts are given as a function of the ratio of the initial defect concentrations c_{0i} calculated after [25]. It is obvious that for corresponding values of both the independent variables the measured temperature shifts are smaller than the calculated ones for $n = 2$. This would mean that either the reaction is lower than of second order or the measured relative resistance recoveries are larger than the medium relative concentration changes thereby producing second-order kinetics only apparently.

A reaction between first and second order is to be expected if the recovery results from a simultaneous migration of defects to their complementary partners and to unsaturable sinks, respectively. When interpreting the temperature shift as a function of the resistance ratio one has, on the other hand, to keep in mind that the mean defect concentration in this case does not increase propor-

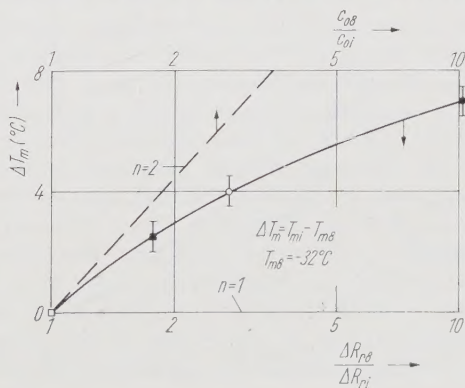


Fig. 4. Temperature shift of peak position versus relative resistance recovery and relative initial defect concentrations, respectively (n reaction order)

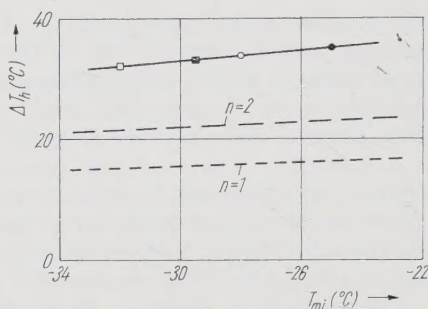


Fig. 5. 'Half-width' of the recovery peak versus peak position (n reaction order)

tionally to the resistance change. Upon increasing the dose a zone nearly saturated with defects is rather likely to be pushed deeper and deeper into the foil. The resistance will thus increase considerably but the mean defect concentration will increase only slightly. In addition, the measurement of the integral concentrations becomes more accurate with increasing mean depth of defect distribution because the undamaged layer, parallel to the damaged layer on top of it, becomes thinner. For that reason the measured relative resistance recovery is also too small. The effect last mentioned is thought to produce the observed deviation from a straight line corresponding to $n = \text{const}$ (Fig. 4). Both influences on resistance measurement are sufficient to cause the deviation from the temperature shift calculated for $n = 2$. Thus, the latter results also show that in the spectra of Fig. 3 the recovery in stage III is scarcely influenced by the defect clusters produced during irradiation.

From the data for the recovery peak (E , T_m , and c_0/c_m) and with the help of the common reaction equations [25] one may calculate a 'half-width', i.e. the temperature difference ΔT_h of the points where the reaction rate attains half its maximum value. Fig. 5 shows ΔT_h as a function of T_m with the order of reaction n as a parameter. For comparison the 'half-widths' found from the recovery curves in Fig. 3 are given after rough background correction. The latter are remarkably broader than the values calculated for homogeneous defect distribution. The deviations from the curve for $n = 2$ thus reflect the expected inhomogeneity of defect distribution after ion irradiation.

The recovery spectra of Fig. 3 agree among others in showing a reaction rate monotonically decreasing with temperature above its pronounced maximum value. This behaviour changes considerably if repeated recovery studies are performed upon low- and medium-dose bombardment after high-dose pretreatment ($\Phi_p > 10^{15} \text{ Au}^+/\text{cm}^2$). A comparison of such spectra (No. 3, 6, and 7 in Fig. 6) with the former (No. 6, 5, and 2 in Fig. 3) shows obvious differences. The maximum of reaction rate is shifted towards lower temperatures and above about -10°C at least one new recovery peak appears. The recovery below -100°C seems to show more structure and the peak at -125°C is more pronounced. Because of the high dose in No. 2 one expects in the sequence of annealing studies shown in Fig. 6 that the spectra were influenced by the high density of the defect clusters. The latter may act as sinks for vacancies and interstitials notwithstanding their type so that the rate of recovery should change in all temperature regions where defects are freely mobile. This might be expected, e.g., in the region of the pronounced peak where, according to the preceding statements, migration and recombination of point defects is likely

Fig. 6. Isochronal recovery spectra for lowered irradiation doses

to occur. In the presence of additional sinks, part of the immobile complementary point defects, otherwise acting as reaction partners, are thought to exist until they also become mobile and precipitate at sinks as the temperature increases. Since in recovery studies after quenching the migration of single and multiple vacancies has always been observed above 0 °C for comparable annealing programs [2, 26 to 28], it is reasonable to explain the recovery peaks observed above about -10 °C also by vacancy migration. The pronounced peak below -10 °C would then result from the recombination of mobile interstitials with vacancies.

This interpretation is supported by comparative investigations on the resistance recovery after different pretreatments, Φ_p , and constant irradiation dose, Φ_i (Fig. 7). The temperature shift of the peak position and the decrease in peak height are quite obvious. This observation confirms the assumption that the recovery below and above about -10 °C is caused by the migration of complementary defects, i.e. interstitials and vacancies, respectively. The effects mentioned are quite evident if a spectrum is taken after normal high-dose pretreatment and intermediate medium-dose irradiation with subsequent annealing at 0 °C. As expected the recovery above 0 °C is quite pronounced in this case.

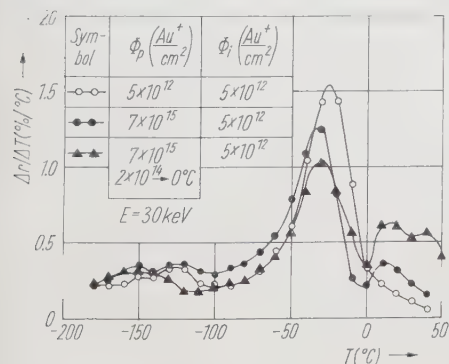


Fig. 7. Normalized fractional recovery for different preirradiations and constant irradiation dose

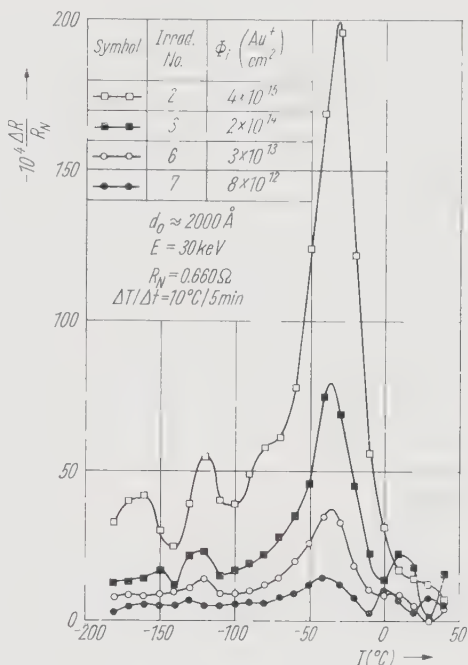


Fig. 8. Normalized fractional recovery for different preirradiations, irradiation energies, and doses

Some further comparative investigations are given to show the influence of irradiation energy on the recovery spectra. Fig. 8 gives three spectra after bombardment with 30, 12, and 5 keV, respectively. It is astonishing at a first glance that, in spite of the comparatively low 30 keV pretreatment, the well-known peak around 20 °C appears for 12 keV. This effect is caused by the short range of 12 keV gold ions in gold [29]. Projectiles with this energy produce defects in a depth where there is a high cluster density resulting from the pretreatment. Besides, the number of primary defects is smaller for this lower energy. After disappearance of the interstitials a measurable part of the vacancies remains and produces the small peak afterwards. Upon 5 keV bombardment the defect production is even smaller than for 12 or 30 keV [22] so that sputtering and cluster production may compete with one another. In this case there is no indication of an additional recovery peak above 0 °C because the 70 Å layer sputtered by the 5 keV bombardment is approximately three times the 'amorphous' range of 12 keV gold ions in gold [29]. The cluster-rich regions near the surface before the 5 keV irradiation are thus likely to be removed. Clusters acting as sinks are, however, necessary in high density to produce a peak above 0 °C. This is not observed, in agreement with the preceding arguments. The differences observed in the temperature region -180 °C to -80 °C are presumably caused by the energy dependence of cascade formation and structure and the spatial distribution of point defects.

In order to complete the investigation, recovery spectra for aligned and random bombardment are compared (Fig. 9). In case of random incidence the recovery is very small above 0 °C. This is probably caused by the very effective spontaneous recombination resulting from the low depth of the defects and the high dose. This assumption is confirmed by considering the fractional resistance recovery, $\Delta R_r/\Delta R_i$. ΔR_i is the total resistance change induced by irradiation. ΔR_r is the fraction recovered up to 40 °C. We were able to plot our results in one diagram by using the square root of the absolute value of the difference $\Delta\Phi = \Phi_i - \Phi_p$ as the independent variable. The shape of the curve (Fig. 10) is likely to be determined by the fact that the resistance change $\Delta R_i - \Delta R_r$ remaining is not only given by the clusters produced in situ but also by those clusters generated during annealing. For the latter the creation probability increases with dose so that $\Delta R_i - \Delta R_r$ increases, i.e. $\Delta R_r/\Delta R_i$ decreases. After high-dose pretreatment the fractional resistance recovery approaches the saturation value of 1.0 for large negative dose differences. This observation

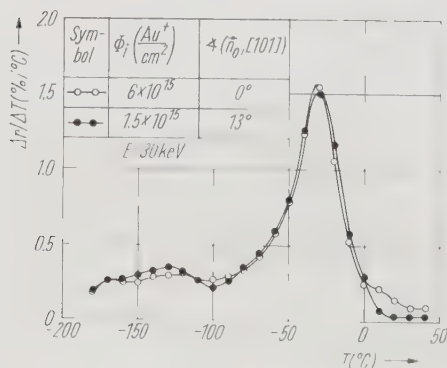
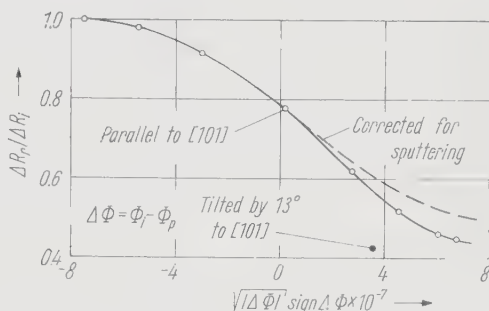


Fig. 9. Normalized fractional recovery for different foil orientation

Fig. 10. Fractional total recovery versus the function of dose difference indicated



is probably caused by the fact that the cluster density equals the saturation value for a considerable depth. Subsequent low-dose bombardment is then thought to produce only a certain rearrangement of clusters.

4. Discussion

For comparison with the results of this paper only equilibrium data and those recovery studies of other authors on gold carried out by resistance measurements after irradiation are used. Electron microscope investigations on interstitial migration in gold [6] are not taken into account because their interpretation remains unsatisfactory until the range of replacement collision sequences is well established.

Dworschak et al. [30] investigated recovery spectra after irradiation with 10 MeV protons at 80 °K. They found that the reaction in stage III follows second-order kinetics with an activation energy of (0.80 ± 0.04) eV. The same result was obtained after 2 MeV electron irradiation at 80 °K for the last 55% of stage III by Bauer and Sosin [31] who thought the recovery to be caused by interstitial migration and recombination with vacancies. Ruhbaum [32] observed the isochronal recovery after 5.3 MeV α -irradiation at 77 up to 470 °K as a function of dose. The results resemble those of the present paper. The spectra for low dose ($\Delta \rho / \rho_{77} < 10\%$) correspond to those with a low pretreatment dosage and the spectra for high dosage ($> 50\%$) match those strongly pretreated. Ermert et al. [33] interpreted the occurrence of 3 MeV α -radiation-induced self-diffusion in the temperature range -60 °C to 10 °C as migration of interstitials. Burger et al. suggest that the recovery found after fast neutron irradiation at 4.6 °K in the so-called stages III_A and III_B is caused by migration of interstitials and recombination with vacancies inside and outside the centres of the defect cascades (peak position of stage III_A: -40 °C).

The above mentioned results are in favour of interstitial migration in stage III with an activation energy of about 0.80 eV. This value is markedly lower than the migration energy of a vacancy, $E_{1V}^M = (0.89 \pm 0.03)$ eV, found from an analysis of equilibrium measurements [4]. Some new investigations, however, produced reasonable doubts as to the consistency of the above mentioned results. Koehler et al. found in very pure samples an activation energy of (0.90 ± 0.04) eV for stage III recovery after quenching, plastic deformation, and 3 MeV electron irradiation, respectively [26 to 28]. They interpreted this value to be the migration energy of a single vacancy.

The results of this paper are now compared to these contradictory investigations and interpretations. Because of the extremely high defect concentrations produced by ion irradiation one may first of all exclude any marked influence of impurities on stage III recovery. The activation energies found make it quite questionable on the other hand to attribute the pronounced recovery peak to the migration of vacancies. This would also disagree with the fact that for high sink densities an additional peak is observed above about 0 °C which, in this case, may only be produced by migration of vacancies or small defect clusters. The latter should be present even in case of low sink density. This is not observed, however. Within the limits of experimental error in the determination of activation energies, it can not be ruled out at a first glance that divacancies ($E_{2V}^M \approx 0.71$ eV [26]) are responsible for the pronounced peak. In that case one would expect, among other things, a migration of divacancies to interstitials or single vacancies. The recovery spectra should then show an additional peak due to the mobility of single or trivacancies. However, this was not found in the samples pretreated with low dose. Finally one has to take into account the formation of di-interstitials due to the high defect density and the possible mobility of interstitials at the irradiation temperature. The chance for this to happen is very low in the case of electron irradiation. Nevertheless, the same activation energy as in this investigation was then found in stage III [31] and ascribed to interstitial mobility. If considerable mobility of multiple defects is excluded, only interstitial migration remains as an acceptable explanation of stage III recovery below about 0 °C. All results of this paper are consistent with such an interpretation. The onset of noticeable interstitial mobility is not found below -80 °C. The activation energy for interstitial migration is $E_{1I}^M = (0.77 \pm 0.07)$ eV.

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